

Broadband single-photon-level memory in a hollow-core photonic crystal fibre

M. R. Sprague^{1*}, P. S. Michelberger¹, T. F. M. Champion¹, D. G. England^{1†}, J. Nunn¹, X.-M. Jin^{1,2}, W. S. Kolthammer¹, A. Abdolvand³, P. St. J. Russell³ and I. A. Walmsley¹

Storing information encoded in light is critical for realizing optical buffers for all-optical signal processing^{1,2} and quantum memories for quantum information processing^{3,4}. These proposals require efficient interaction between atoms and a well-defined optical mode. Photonic crystal fibres can enhance light-matter interactions and have engendered a broad range of nonlinear effects⁵; however, the storage of light has proven elusive. Here, we report the first demonstration of an optical memory in a hollow-core photonic crystal fibre. We store gigahertz-bandwidth light in the hyperfine coherence of caesium atoms at room temperature using a far-detuned Raman interaction. We demonstrate a signal-to-noise ratio of $2.6:1$ at the single-photon level and a memory efficiency of $27 \pm 1\%$. Our results demonstrate the potential of a room-temperature fibre-integrated optical memory for implementing local nodes of quantum information networks.

Non-classical light can possess a complex set of correlations. These correlations can be exploited in order to simulate quantum systems⁶, make precise measurements⁷ or perform efficient computation⁸. However, the generation of these states often relies on probabilistic sources where the probability of generating, for instance, N heralded single photons scales exponentially in N . A potential solution is to use a quantum memory—a device that faithfully stores non-classical states of light—as a temporal multiplexer to synchronize heralded, probabilistic events^{3,9}. With a memory, the success rate of multiphoton generation, or entanglement generation, can be enhanced by base factor Γ , which is the product of the time-bandwidth product T (the ratio of the memory lifetime to the pulse duration) and the memory efficiency η (ref. 9). Creating non-classical multiphoton states therefore requires an array of broadband, efficient quantum memories.

Efficient light-matter coupling can be achieved with optical cavities¹⁰ or ensembles of atoms^{11,12}. For ensembles, the interaction can be enhanced by confining the atoms and optical mode in a waveguide. Waveguide structures afford tight transverse confinement of optical modes over a long length, limited only by the attenuation in the waveguide. This allows for stronger light-matter coupling than is possible with free-space beams and bulk ensembles, where the product of the intensity of the mode and the Rayleigh length is constant, limited by diffraction. The lower power requirement in a waveguide reduces the degree of filtering needed to isolate the weak signal from the strong control field. It also enables larger-scale operation; multiple memories can be operated with the same power required for a single bulk memory. Nonlinear optical effects have been investigated in a variety of integrated structures including antiresonant reflection optical waveguides¹³, evanescently coupled optical fibres¹⁴ and hollow-core photonic-bandgap fibres

(HC-PBGFs)¹⁵ loaded with alkali atoms. HC-PBGFs, in particular, are attractive because of their low transmission loss. Cross-phase modulation¹⁶, electromagnetically induced transparency¹⁷ and four-wave mixing¹⁸ have been realized in an HC-PBGF with a $6\text{-}\mu\text{m}$ -diameter core.

The storage of light in a HC-PBGF has not been realized, however, because of two main obstacles. First, the small optical mode causes significant transit-time broadening at room temperature, and the memory interaction must occur within the time that the atoms take to traverse the guided mode. Second, it is challenging to load a sufficient number of atoms into the core to achieve the high optical depth required for efficient light storage. Both obstacles can be surmounted by using large-core kagome-structured hollow-core photonic crystal fibres (HC-PCFs) to increase the transit time and the rate at which atoms are loaded into the fibre core^{19,20}, and by using a far-off-resonant, broadband Raman interaction^{21–23} to perform the memory protocol within the transit time.

Here, we demonstrate light storage in a $26\text{-}\mu\text{m}$ -diameter core kagome-structured HC-PCF loaded with caesium atoms at room temperature, as illustrated in Fig. 1a. Large-core kagome HC-PCFs predominantly support the fundamental spatial mode with low loss (1.5 dB m^{-1} at 852 nm) given careful mode matching²⁴. A 20-cm -long fibre was mounted in an ultrahigh-vacuum system and passively loaded with caesium atoms (released from a glass ampoule)¹⁹. A schematic of the experiment is shown in Fig. 1b. The optical depth was first increased by an order of magnitude (to on the order of several thousand) using light-induced atomic desorption (LIAD) to transiently desorb atoms from the surface of the fibre^{19,25}. The atoms were then optically pumped into the $6^2\text{S}_{1/2}(F=4)$ ground state in preparation for the memory interaction¹⁹.

The signal and control pulses, separated in frequency by 9.2 GHz , were launched, orthogonally polarized, into the fibre. The Raman interaction stored the weak signal field in a spin coherence between the dipole-decoupled storage and ground states, the $6^2\text{S}_{1/2}(F=3,4)$ hyperfine levels, via a two-photon transition with a strong control field (Fig. 1c). Both the signal and control fields were detuned by 15.2 GHz from an intermediary excited state, the $6^2\text{P}_{3/2}$ excited-state hyperfine manifold. After a programmable delay, a second control pulse mapped the spin coherence into a retrieved optical mode. The retrieved signal was filtered from the control field using polarization and frequency degrees of freedom and then detected with an avalanche photodiode (APD).

Figure 2a shows the storage and retrieval of a bright signal pulse comprising $\sim 1 \times 10^6$ photons. When no control field is present, the signal is transmitted through the atomic vapour. When the control pulse is applied in the read-in time bin at $t = 0$ the signal is absorbed

¹Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, UK, ²Department of Physics, Shanghai Jiao Tong University, Shanghai 200240, China, ³Max Planck Institute for the Science of Light, 91058 Erlangen, Germany, [†]Present address: National Research Council of Canada, 1200 Montreal Road, Ottawa, Ontario K1A 0R6, Canada. *e-mail: michael.sprague@physics.ox.ac.uk

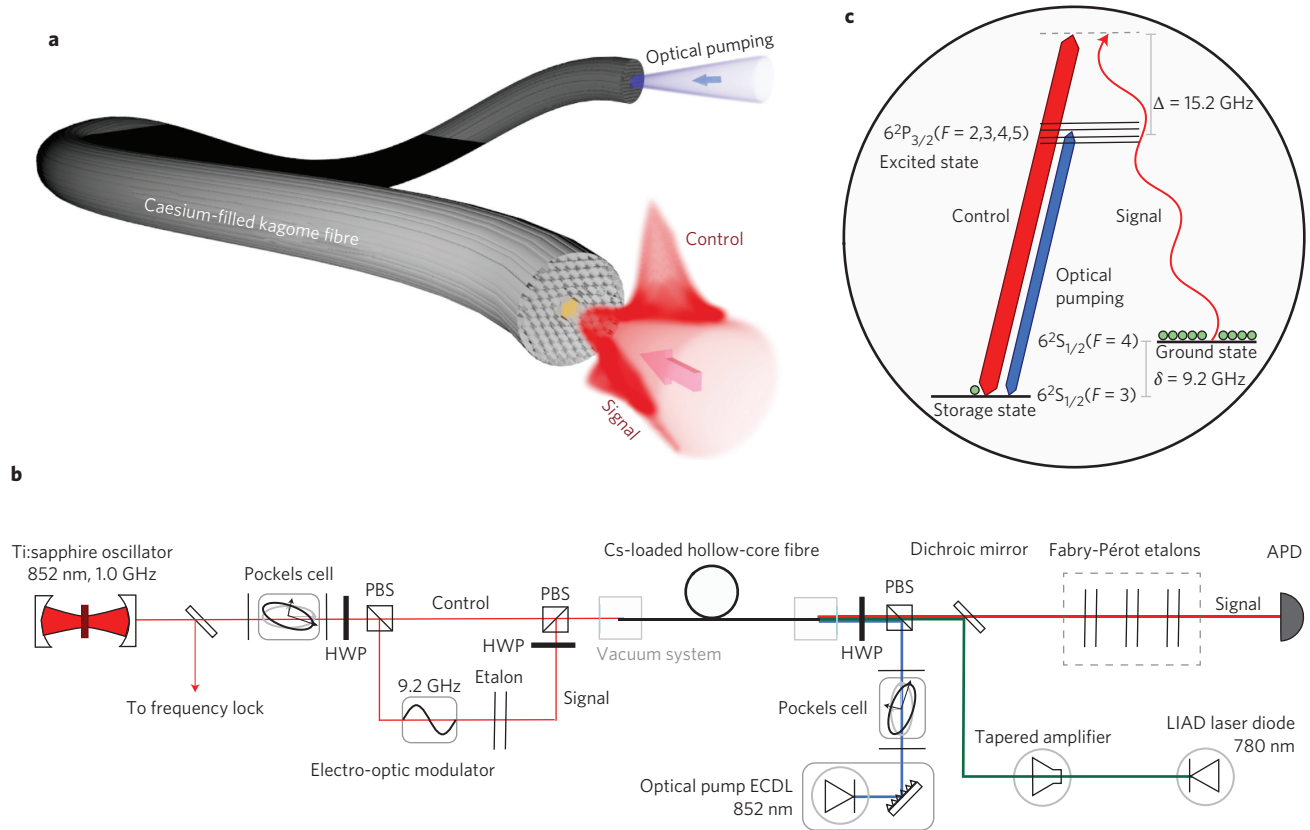


Figure 1 | Raman quantum memory in kagome hollow-core fibre. **a**, Illustration of the caesium-filled kagome fibre showing control and signal pulses. **b**, Experimental set-up (see Methods; HWP, half-wave plate; PBS, polarizing beamsplitter; ECDL, external-cavity diode laser; APD, avalanche photodiode). The signal field was derived from the same laser using an electro-optic modulator driven at 9.2 GHz and an etalon to isolate the red sideband. **c**, Three-level Λ -system showing signal and control field in two-photon resonance detuned from the D_2 excited-state manifold by 15.2 GHz. The control and signal pulses had a bandwidth of 1.0 GHz at a centre wavelength of 852 nm. The optical pumping beam was switched off with a Pockels cell 5 ns before memory interaction. Green circles represent the population of caesium atoms in the hyperfine levels.

into the memory, and at $t = 12.5$ ns a second control pulse retrieves the signal in the read-out time bin. The pulse sequence for the measurements is illustrated in Fig. 2b. The memory efficiency was calculated from the ratio of the integrated intensity of the retrieved and transmitted signals with a 5 ns window (see Supplementary Section 'Calculation of memory efficiency'). The total memory efficiency was $27 \pm 1\%$ for a control pulse energy of 40 pJ, which is a 200-fold reduction in energy compared to a free-space memory²³.

We now examine how the memory efficiency depends on the Raman coupling. The Raman coupling constant C^2 defines the mixing between the optical and atomic modes and is given by $C^2 \approx d(\gamma/\delta)(\Omega/\Delta)^2$, where γ is the homogeneous linewidth of the excited state, δ is the bandwidth of the pulse to be stored, Ω is the Rabi frequency of the control field, Δ is the excited-state detuning and d is the resonant optical depth²¹. The optical depth in the fibre was varied dynamically using LIAD while keeping Ω constant. The optical depth was measured from the delay experienced by signal pulses in the absence of a control pulse. The dispersion of the pulse was then related to the optical depth via a Kramers–Kronig relation (see Supplementary Section 'Estimation of optical depth'). These measurements were interleaved with the memory interaction to simultaneously record the optical depth and memory efficiency, which are plotted in Fig. 2c together with the Raman coupling constant.

The results were compared to an adiabatic, linear, one-dimensional model of the Raman interaction²¹. We optimized over Ω as the only fit parameter, giving an estimate of 2.8 ± 0.1 GHz,

compared to an experimentally estimated mean Rabi frequency of 3.4 ± 0.5 GHz. This reasonable agreement shows that the model faithfully describes the memory interaction in the hollow-core fibre. The memory efficiency is currently limited by the Raman coupling and, ultimately, limited to $\sim 50\%$ by the spatial mode mismatch between the stored spin mode and the read-out control pulse²¹. Higher efficiency could be reached by either increasing the integrated intensity of the read-out control pulse or reading out the memory in the backward direction. The control field would need to be temporally stretched to increase the total pulse energy while keeping the peak intensity low enough to respect the adiabatic condition. Efficient backward retrieval requires the length of the ensemble to be restricted such that the product of the wavevector mismatch and length is much less than one. In principle, the memory efficiency could approach unity when the Raman coupling constant is much greater than one²¹.

We now demonstrate storage at the single-photon level (Fig. 3a). We coupled a coherent-state signal pulse with an average of 1.8 ± 0.1 photons per pulse into the fibre for storage. The read-in time bin shows Raman absorption and the read-out time bin retrieval when the memory interaction is turned on. The inset of Fig. 3a shows the signal retrieved when no signal is sent into the memory; this is the unconditional noise level of the memory. The signal-to-noise ratio (SNR) at the single-photon level as a function of memory efficiency is plotted in Fig. 3b, normalized for an input of strictly one photon per pulse, giving a maximum value of 2.6. The noise level was calculated from the noise generated by another control pulse separated by 50 ns from the memory interaction.

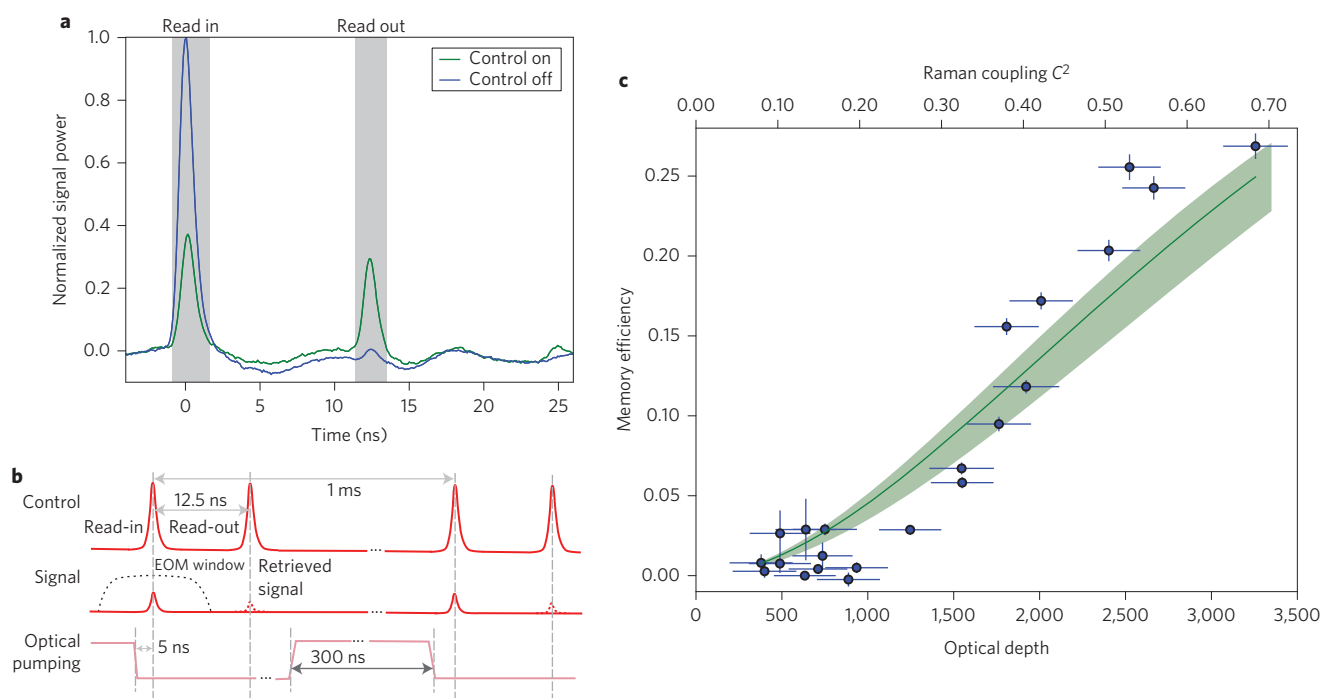


Figure 2 | Storage of classical light in a kagome hollow-core fibre. **a**, Time-domain trace showing the signal consisting of 1×10^6 photons with (green line) and without (blue line) the control pulse applied in the read-in and read-out time bins (grey shading). The finite response time of the electro-optic modulator caused a small amount of signal in the read-out time bin when the control field was not present; this signal was subtracted from the estimate of the memory efficiency. **b**, Pulse sequence for classical and single-photon-level measurements. The LIAD field remained on for the duration of the measurements. **c**, The dependence of memory efficiency on optical depth fitted to a theoretical model of the Raman interaction²¹ (green line), with a one-standard-deviation confidence interval predicted from nonlinear regression (green shading). The corresponding Raman coupling constant C^2 is shown on the upper x-axis. Errors bars represent the uncertainty in pulse delay (horizontal bars) and signal pulse energy (vertical bars).

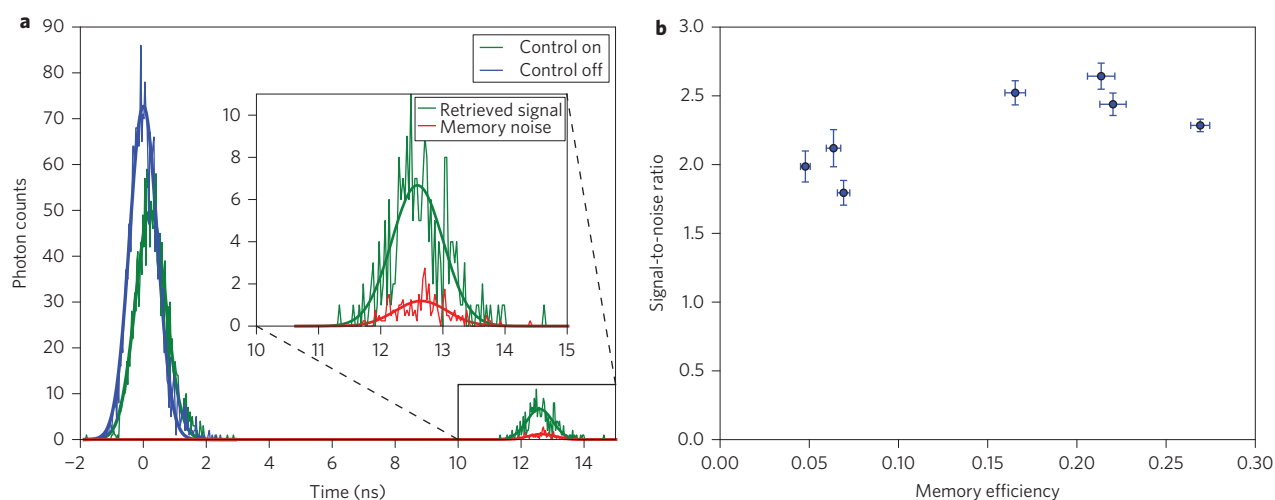


Figure 3 | Storage of single-photon-level pulses. **a**, Time-domain histogram reconstructed from the single-photon APD and time-to-amplitude converter, fit to a Gaussian function (thick line). The signal is shown with (green line) and without (blue line) the control field applied. Inset: unconditional noise (red line) in the read-out time bin. **b**, SNR at the single-photon level as a function of memory efficiency (normalized to one input photon per pulse). Error bars are estimated from the standard deviation of the count statistics.

Any residual coherence could seed the noise process, so the estimate of the SNR represents a lower bound. However, as discussed below, the memory lifetime was less than 50 ns.

The noise was likely caused by spontaneous four-wave mixing (SFWM)²³; the control pulse can act on the ground state, in addition to the storage state, scattering an anti-Stokes photon and creating a spurious spin excitation that can then be read out from the storage state by a second interaction with the control pulse. The noise was in

the same spatial, temporal and frequency mode as the retrieved signal. In addition, the SNR was approximately independent of the memory efficiency, indicating that the memory retrieval and noise generation had the same scaling with C^2 . This is consistent with SFWM, as opposed to imperfect control field filtering, because the control field power was kept constant throughout the measurements and only the optical depth was varied. The SNR could potentially be improved by reducing the detuning or

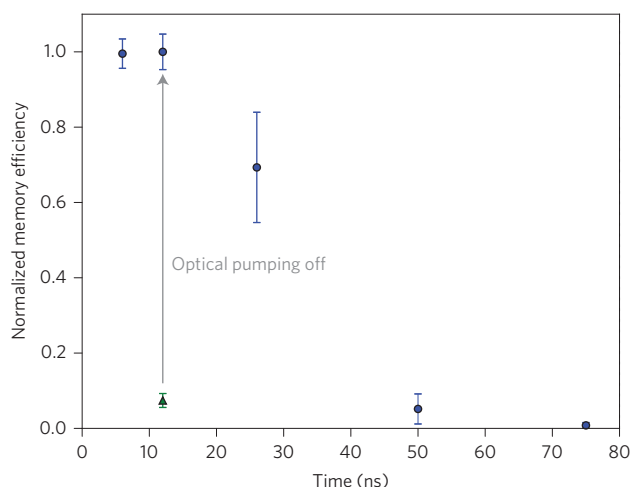


Figure 4 | Lifetime of memory in hollow-core fibre. Memory efficiency as delay between read in and read out increased. The memory efficiency was normalized to the memory efficiency measured at 6 ns. An unbalanced, fibre-based Mach-Zehnder interferometer generated two control pulses with variable delay between storage and retrieval. The triangular point shows the memory efficiency at 12.5 ns when the optical pumping was left on. Error bars are the standard deviation of repeated measurements.

selectively filtering anti-Stokes photons in the memory using a fibre-integrated Fabry-Pérot etalon²⁶.

The single-photon-level SNR is, to our knowledge, the highest yet demonstrated for a room-temperature memory with an atomic vapour²³. To interface the memory with a heralded single-photon source, the heralding efficiency should exceed the inverse of the SNR to enable non-classical operation. This requires a minimum 40% heralding efficiency, which is feasible given current heralded single-photon sources²⁷. Furthermore, we note that the noise is sufficiently low that a Bell violation could be achieved by storing and retrieving one half of a Bell pair²³.

The lifetime of the memory is shown in Fig. 4, which was measured at an optical depth of 2,500. The measured lifetime ($1/e$) is 27 ± 9 ns, although we note that the shape indicates that the decoherence mechanism is non-Markovian. Figure 4 shows that the memory efficiency decreased by an order of magnitude when the optical pumping was left on during the memory interaction (it was switched off for Figs 2 and 3). The optical pumping beam spoiled the spin coherence by coupling the storage and excited state, leading to spontaneous decay and a spatially dependent Stark shift. The expected decoherence induced by the transit time of the atoms across the fibre mode predicts a lifetime ($1/e$) of 100 ns. We attribute this discrepancy to residual transmission of the optical pumping beam, a technical limitation of the Pockels cell. The memory lifetime could be further extended by using a buffer gas or larger-diameter core to increase the transit time and applying a spin-preserving coating to reduce the decoherence rate due to wall collisions²⁸. We note that, given the current lifetime, the memory is suitable for the synchronization of single-photon sources. The time-bandwidth product T is already 100 and could result in a base factor enhancement $\Gamma = \eta T = 30$ in the generation of two synchronized single photons⁹.

In conclusion, we have demonstrated, for the first time, the storage of light in an HC-PCF. The low noise at the single-photon level (0.1 noise photons/pulse for $\eta = 27\%$) and the high time-bandwidth product underpins the memory's potential for synchronizing heralded single-photon sources for local networks. The fibre memory requires 200 times less pulse energy than a free-space Raman memory, which could enable the operation of multiple memory units using a single control laser. Frequency

conversion or dual-frequency single-photon sources could be used to integrate the memory with telecommunication-wavelength optical systems²⁹. Finally, splicing the kagome fibre to a single-mode fibre³⁰ would realize a compact, integrated quantum memory at room temperature.

Methods

Oscillator, optical pumping and LIAD. The oscillator was a mode-locked Ti:sapphire laser fitted with an intracavity Gires-Tournois interferometer to produce a train of 365 ps pulses (full-width at half-maximum, FWHM) at a repetition rate of 80 MHz and a centre wavelength of 852 nm. The pulse repetition rate was reduced to between 1 and 8 kHz using a Pockels cell triggered by the oscillator. The Pockels cell had an extinction ratio of 3,000:1. The signal field was generated with an electro-optic modulator (EOM) driven at 9.2 GHz and 38.9 GHz free-spectral-range (FSR) etalon in order to isolate a red-detuned sideband. For classical and single-photon-level storage, the control field energy per pulse coupled into the fibre was 40 pJ, resulting in a peak power of ~ 100 mW. The optical pumping beam was derived from an external cavity diode laser (ECDL) locked to the $6^2S_{1/2}(F=3)$ to $6^2P_{3/2}(F'=3)$ transition. The beam was switched on for 300 ns before memory interaction and switched off 5 ns before the signal and control were launched into the fibre. The beam used for LIAD was derived from an ECDL operating at 780 nm. The laser beam was amplified using a tapered amplifier and launched counter-propagating, using a dichroic mirror, into the HCF with an in-coupled power of 75 mW. See Supplementary Sections 'Oscillator for signal and control fields' and 'Optical pumping and light-induced atomic desorption' for more details.

Hollow-core photonic crystal fibre and vacuum system. The HC-PCF has a kagome structure, formed by a trigonal lattice with a set of defects that also forms a trigonal lattice, but with double the period. The fibre does not have a bandgap, but guides via a mechanism related to antiresonant reflection²⁴. The fibre loss was 1.5 dB m^{-1} at 852 nm and the coupling efficiency into the fibre was 65%. The coupling efficiency of the mode emerging from the HC-PCF into a standard single-mode fibre was 70%. The fibre was found to be polarization-maintaining. The polarization extinction ratio after transmission through the HC-PCF and coupling to a single-mode fibre, using cross-polarizers, was greater than 4,000:1.

Detection and filtering. The combined polarization filtering had an extinction ratio greater than 1×10^{-3} . The frequency filtering was realized with a set of three planar air-gapped etalons with an FSR of 18 GHz and the attenuation between control and signal modes was 1×10^{-2} for each etalon. For the single-photon-level measurements, a 10^3 GHz FSR air-gapped etalon was used in a double-pass configuration to eliminate the anti-Stokes sideband. For the single-photon-level measurements, the signals in the read-in and read-out time bins were recorded with a field-programmable gate array and, simultaneously, with a time-to-amplitude converter in conjunction with a multichannel analyser.

Received 20 September 2013; accepted 4 February 2014;
published online 16 March 2014

References

- Ramaswami, R., Sivarajan, K. & Sasaki, G. *Optical Networks: A Practical Perspective* (Morgan Kaufmann, 2009).
- Zhu, Z., Gauthier, D. J. & Boyd, R. W. Stored light in an optical fiber via stimulated Brillouin scattering. *Science* **318**, 1748–1750 (2007).
- Sangouard, N., Simon, C., de Riedmatten, H. & Gisin, N. Quantum repeaters based on atomic ensembles and linear optics. *Rev. Mod. Phys.* **83**, 33–80 (2011).
- Lukin, M. D. Trapping and manipulating photon states in atomic ensembles. *Rev. Mod. Phys.* **75**, 457–472 (2003).
- Russell, P. S. Photonic-crystal fibers. *J. Lightwave Technol.* **24**, 4729–4749 (2006).
- Aspuru-Guzik, A. & Walther, P. Photonic quantum simulators. *Nature Phys.* **8**, 285–291 (2012).
- Giovannetti, V., Lloyd, S. & Maccone, L. Advances in quantum metrology. *Nature Photon.* **5**, 222–229 (2011).
- Pan, J.-W. *et al.* Multiphoton entanglement and interferometry. *Rev. Mod. Phys.* **84**, 777–838 (2012).
- Nunn, J. *et al.* Enhancing multiphoton rates with quantum memories. *Phys. Rev. Lett.* **110**, 133601 (2013).
- Specht, H. P. *et al.* A single-atom quantum memory. *Nature* **473**, 190–193 (2011).
- Julsgaard, B., Sherson, J., Cirac, J. I., Fiurasek, J. & Polzik, E. S. Experimental demonstration of quantum memory for light. *Nature* **432**, 482–486 (2004).
- Choi, K. S., Deng, H., Laurat, J. & Kimble, H. J. Mapping photonic entanglement into and out of a quantum memory. *Nature* **452**, 67–71 (2008).
- Wu, B. *et al.* Slow light on a chip via atomic quantum state control. *Nature Photon.* **4**, 776–779 (2010).
- Spillane, S. *et al.* Observation of nonlinear optical interactions of ultralow levels of light in a tapered optical nanofiber embedded in a hot rubidium vapor. *Phys. Rev. Lett.* **100**, 233602 (2008).

15. Cregan, R. F. *et al.* Single-mode photonic band gap guidance of light in air. *Science* **285**, 1537–1539 (1999).
16. Venkataraman, V., Saha, K. & Gaeta, A. L. Phase modulation at the few-photon level for weak-nonlinearity-based quantum computing. *Nature Photon.* **7**, 138–141 (2013).
17. Ghosh, S. *et al.* Low-light-level optical interactions with rubidium vapor in a photonic band-gap fiber. *Phys. Rev. Lett.* **97**, 023603 (2006).
18. Londero, P., Venkataraman, V., Bhagwat, A. R., Slepkov, A. D. & Gaeta, A. L. Ultralow-power four-wave mixing with Rb in a hollow-core photonic band-gap fiber. *Phys. Rev. Lett.* **103**, 043602 (2009).
19. Sprague, M. R. *et al.* Efficient optical pumping and high optical depth in a hollow-core photonic-crystal fibre for a broadband quantum memory. *New J. Phys.* **15**, 055013 (2013).
20. Perrella, C., Light, P. S., Stace, T. M., Benabid, F. & Luiten, A. N. High-resolution optical spectroscopy in a hollow-core photonic crystal fiber. *Phys. Rev. A* **85**, 012518 (2012).
21. Nunn, J. *et al.* Mapping broadband single-photon wave packets into an atomic memory. *Phys. Rev. A* **75**, 011401 (2007).
22. Reim, K. F. *et al.* Towards high-speed optical quantum memories. *Nature Photon.* **4**, 218–221 (2010).
23. Reim, K. F. *et al.* Single-photon-level quantum memory at room temperature. *Phys. Rev. Lett.* **107**, 053603 (2011).
24. Benabid, F. & Roberts, P. Linear and nonlinear optical properties of hollow core photonic crystal fiber. *J. Mod. Opt.* **58**, 87–124 (2011).
25. Slepkov, A. D., Bhagwat, A. R., Venkataraman, V., Londero, P. & Gaeta, A. L. Generation of large alkali vapor densities inside bare hollow-core photonic band-gap fibers. *Opt. Express* **16**, 18976–18983 (2008).
26. Wang, X., Zhu, T., Chen, L. & Bao, X. Tunable Fabry-Perot filter using hollow-core photonic bandgap fiber and micro-fiber for a narrow-linewidth laser. *Opt. Express* **19**, 9617–9625 (2011).
27. Krapick, S. *et al.* An efficient integrated two-color source for heralded single photons. *New J. Phys.* **15**, 033010 (2013).
28. Bradley, T., McFerran, J. J., Jouin, J., Thomas, P. & Benabid, F. in *OSA Technical Digest*, CM3I.2 (Optical Society of America, 2013).
29. Fernandez-Gonzalvo, X. *et al.* Quantum frequency conversion of quantum memory compatible photons to telecommunication wavelengths. *Opt. Express* **21**, 19473–19487 (2013).
30. Benabid, F., Couny, F., Knight, J. C., Birks, T. A. & Russell, P. S. J. Compact, stable and efficient all-fibre gas cells using hollow-core photonic crystal fibres. *Nature* **434**, 488–491 (2005).

Acknowledgements

The authors thank D. Saunders for comments on the manuscript. The work was supported by the Engineering and Physical Sciences Research Council (EPSRC; EP/J000051/1), the Quantum Interfaces, Sensors, and Communication based on Entanglement Integrating Project (EU IP Q-ESSENCE; 248095), the Air Force Office of Scientific Research: European Office of Aerospace Research & Development (AFOSR EOARD; FA8655-09-1-3020), EU IP SIQS (600645), the Royal Society, the Clarendon Fund (to M.R.S.), St Edmund Hall (to M.R.S.), EU ITN FASTQUAST (to P.S.M.), and an EU Marie-Curie Fellowship (PIIF-GA-2011-300820 to X.-M.J.; PIEF-GA-2012-331859 to W.S.K.).

Author contributions

M.R.S. designed the experiment and built it with assistance from D.G.E., P.S.M., W.S.K. and T.F.M.C. A.A. and P.St.J.R. designed and drew the fibre and provided valuable insights. M.R.S. collected and analysed the data. J.N. and M.R.S. performed the comparison to theory. The project was conceived by M.R.S., J.N., X.M.J. and I.A.W. M.R.S. wrote the manuscript with input from all authors.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to M.R.S.

Competing financial interests

The authors declare no competing financial interests.